Calculations of the Thermal Conductivity of NIF Target Materials using Finite-temperature Quantum Molecular Dynamics

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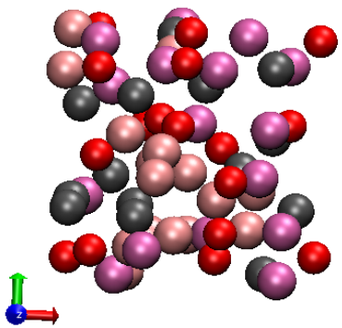
Using finite-temperature density functional theory MD, we performed simulations of several important materials in the Inertial Confinement Fusion-National Ignition Facility nominal target designs, comprising various mixtures of Be, CH, D, and T atoms. Simulations were done over a range of temperatures between 5 eV and 20 eV, at densities between 7.5 and ~14 g/cc. From the MD trajectories, we calculated the electrical and thermal conductivity.

Fig. 1. A molecular dynamics snapshot of a plastic/DT mixture in a 3D periodic cell: contains 14 carbon atoms and 18 hydrogen atoms (plastic constituents) and 16 deuterium and 16 tritium atoms.



The centerpiece of the US Inertial Confinement Fusion (ICF) program is the National Ignition Facility (NIF), located at LLNL. It is anticipated that the NIF will demonstrate thermonuclear ignition in the near future. Initial tests will begin with a hohlraum-driven, spherical, single-shell capsule composed of three principal layers: on the outside, a low-Z ablator (carbon-hydrogen [CH] or beryllium [Be]) that encapsulates a frozen deuterium-tritium (DT) layer containing the DT gas. Energy from the intense laser light, interacting with the inside of the hohlraum, creates a series of strong shock waves that compress and gradually heat the DT fuel to generate the fusion reactions. As the fuel/ablator interface approaches its maximum velocity, densities are expected to be in the vicinity of 10 g/cc with temperatures in the range of 10 to 20 eV [1], and the materials will be highly ionized plasmas. It is at these intermediate times, during the deceleration/compression phase, that the occurrence of hydrodynamic instabilities is most acute. Local ripples at the ablator/fuel interface, inherent in the fabrication process or fed through from the ablation front, can then grow, leading to interpenetration of the ablator material into the fuel (DT)–and the mixing of ablator material with the fuel can degrade performance. The control of the densities of the ablator material and fuel is then critically important to mitigate this instability, which can cause deleterious mixing. The Rayleigh-Taylor instability growth rate becomes larger when the ratio of the ablator density to that of the fuel increases. Since, at constant pressure, the density varies inversely with the temperature, one would expect that the thermal conductivity of the materials would be a key parameter in the optimization of target designs. The thermal conductivity model used (Lee-More [2]) for NIF target design is uncertain in this regime [1], and experimental data is difficult to obtain.

The conditions present in the capsule and ablator, during the compression of the fuel, fall within an area of condensed matter physics termed Warm Dense Matter (WDM). It encompasses various astrophysical phenomena (such as planetary interiors and white dwarf atmospheres), laser-matter interactions, and shock compression and exploding wire experiments. Considerable effort has gone into the development of computational methods to study these plasma systems. The problem is challenging because the material is only partially ionized, requiring that ions, free electrons, and molecules all be treated simultaneously at the quantum mechanical level. The focus of this work is to apply established ab initio computational methods [3] to calculate the thermal conductivity of pure ICF target materials and their mixtures.

Molecular Dynamics (MD) simulations and optical analysis were performed with the Vienna ab initio Simulation Package (VASP)[4-6]. VASP provides a fully quantum mechanical treatment of the electrons by employing a plane-wave, Finite Temperature Density Functional Theory (FTDFT) solution to Schroedinger's equation in the generalized gradient approximation (GGA) of Perdew-Wang 91 [7]. The FTDFT method has been found to predict optical conductivities that are in reasonable agreement with plasma experiments on copper (Cu) [8] and aluminum (Al) [3,9]. Assuming the usual Born-Oppenheimer approximation, the ion trajectories are evolved classically (using a velocity Verlet algorithm) in the isokinetic ensemble from the Density Functional Theory (DFT)-calculated electron densities and the Coulombic ion-ion repulsion. Local thermodynamic equilibrium (LTE) is enforced between the ions and electrons, that is, Ti = Te.

The DFT program produces the Kohn-Sham orbitals, ψ_i, and associated eigenenergies, ε_i, from which we determine both the electrical and thermal conduction properties from a Chester-Tellung-Kubo-Greenwood formulation [10] by calculating various frequency-dependent kinetic coefficients, L_{nm}, using the basic form [9,11]:

$$L_{nm}(\omega) = (-1)^{n+m} \frac{2\pi e^2 \hbar^2}{\Omega m^2} \sum_{i,j} (\varepsilon_i - \mu)^{n-1} (\varepsilon_j - \mu)^{m-1} F_{ij}(\omega) \left| D_{ij} \right|^2 \delta(\varepsilon_i - \varepsilon_j - \hbar\omega)$$

where Ω is the atomic volume, *e* the electrical charge, m the electron

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mass, μ is the chemical potential or Fermi energy, and i and j are summed over Kohn-Sham orbitals. We have omitted the summation over k-points for clarity. F_{ij} represents the difference of two Fermi-Dirac distributions f_{FD} at temperature T and D_{ij} the velocity dipole matrix.

By taking the zero-frequency limit of these quantities, we can recover the real part of the *dc* electrical conductivity: $\sigma_{dc} = L_{11}(0) = \sigma(0)$,

where $\sigma(\omega)$ is the frequency-dependent electrical conductivity. The thermal conductivity κ is:

$$\kappa = \frac{1}{T}\left(L_{22}(0) - \frac{L_{12}^2(0)}{L_{11}(0)} \right).$$

A more detailed description of the procedures for calculating the kinetic coefficients, especially in regards to the electrical conductivity, appears elsewhere [9].

For some physical cases, a simple relationship exists between the electric and thermal conductivities known as the Wiedemann-Franz Law (WF) [10]: $\kappa = \sigma LT$

where T is the temperature and L is a constant (the Lorentz number equal to 2.44 x10^{-8} watt × ohm × deg^{-2}). In some of the regimes, especially for metals, the frequency-dependent electrical conductivity obeys the simple Drude formula [9]:

$$\sigma(\omega) = \frac{\sigma_{dc}}{1 + (\omega\tau)^2},$$

where τ is the collisional relaxation time. Fitting $\sigma(\omega)$ to this form yields σ_{dc} and τ as well as the electron density:

$$n_e = \frac{m\sigma_{dc}}{e^2\tau},$$

and the effective ionization fraction: $\overline{Z} = n_e\Omega/N$, where N is the number of atoms in a simulation cell.

NIF target materials were represented by ensembles of 64 atoms, corresponding to the ablator material (Be or CH), the capsule material (DT, 1:1) and equal mixtures of the ablator and capsule materials (1:1). A constant time step of 0.1 fs was used for all MD simulations. Figure 1 gives a snapshot from a DFT-MD simulation of a mixture of plastic and DT fuel. For MD simulations, the number of electronic energy bands was chosen so that the occupation fraction of the highest band was less than 0.0001 except for the highest energy cases (20 eV), where the number of bands were chosen to yield an occupation fraction of 0.001. This was done for reasons of computational expediency. A test was performed on the CH system to verify that this reduction in the number of bands did not have a significant effect on the calculated conductivity. For the optical analysis, the number of bands used was three times the value used in the MD simulation. Since we found that both the thermal conductivity and $\overline{Z}$ are essentially constant over the temperature range studied, we are justified in taking the averages. These average values are listed in Table 1. Table 1 also lists the number of energy bands used in the MD simulation, the electron density, and the collision frequency τ. Note that the thermal conductivities of the mixed systems are not close to the arithmetic means of the constituent materials. Future work will include calculations of the thermal conductivity for other mixtures of ablator and fuel.

Table 1. Number of bands used in DFT-MD calculation, thermal conductivity, average number of free electrons per atom, electron density, and electron collision time (all at density= 10 g/cc). All entries are for a temperature of 10 eV except the thermal conductivity, which is averaged over all energies.

Material	Number of bands, MD	Thermal Conductivity (kW/K•m)	Z	Electron Density (Å-3)	Tau (10-17 s)
Be	364	2.8	2.2	1.50	2.20
CH	287	5.5	2.6	3.09	2.38
DT	175	17.6	1.1	2.71	8.13
Be/DT	331	3.9	1.8	1.82	2.57
CH/DT	206	7.4	1.8	2.60	3.51

[1] Hammel, B.A., et al., *High Energy Density Physics* **6,** 171 (2010).
[2] Lee, Y.T., and R.M. More, *Phys Fluids* **27,** 1273 (1984).
[3] Horner, D.A., et al., *Phys Rev B* **81** (2010).
[4] Kresse, G., and J. Hafner, *Phys Rev B* **47**, 558 (1993).
[5] Kresse, G., and J. Furthmüller, *Comput Mat Sci* **6,** 15 (1996).
[6] Kresse, G., and J. Furthmüller, *Phys Rev B* **54,** 11169 (1996).
[7] Perdew, J.P., in *Electronic Structure of Solids*, Eds. F. Ziesche and H. Eschrig, Akademie Verlag, Berlin (1991).
[8] Clerouin, J., et al., *Phys Rev B* **71,** page # (2005).
[9] Mazevet, S., et al., *Phys Rev E Stat Nonlin Soft Matter Phys* **71,** 016409 (2005).
[10] Chester, G.V., and A. Thellung, *Proc Phys Soc Lond* **77,** 1005 (1961).
[11] Recoules, V., and J.P. Crocombette, *Phys Rev B* **72**, page # (2005).

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